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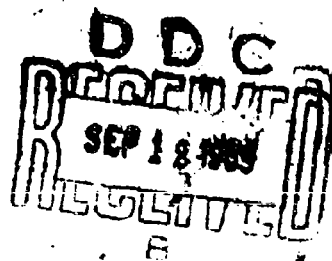# FINAL SCIENTIFIC REPORT

THE INFLUENCE OF INITIAL TEMPERATURE ON THE LIMITS OF DETONABILITY

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Abstract

The influence of initial temperature ($T_o = 195\ ^oK$ and $T_o = 295\ ^oK$) on the limits of detonability of gaseous detonations is investigated for mixtures of $CH_4 - O_2$, $H_2 - O_2$, and $H_2 - $ air at one atmosphere initial pressure in long tubes of 4, 6, 10, 16 and 26 mm inner diameter, respectively, by use of a rotating drum camera. Two experimental designs are described to measure these limits at low initial temperatures. The regime of fuel gas concentrations for stable detonations becomes somewhat narrower at lower initial temperatures. The influence of the initial temperature on the critical tube diameter is estimated. Furthermore the applicability of the Chapman-Jouguet theory is discussed in relation to shock wave configurations, i.e. the Mach triple configuration. The essential arguments for the existence of the limits of detonability are discussed. Finally a new method to estimate the limits of detonability with a very simple experimental arrangement is des-

cribed. By this method experiments concerning

the stability of a detonation and its initiation

process are linked together.

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LISTS OF ABBREVATIONS AND SYMBOLS

D = detonation velocity

$M = \dfrac{D}{a}$ = Mach number

Q = heat of reaction per unit of mass

T = temperature

a = velocity of sound

c = concentration in Vol %

d = tube diameter

p = pressure

$\varkappa$ = ratio of the specific heats

ν = frequency of spin

ρ = density

Inde   CJ   refers to Chapman-Jouguet plane
        S   refers to values behind the front shock wave
        L   refers to the limits of detonability
        o   refers to the undisturbed fresh gas

LIST OF ILLUSTRATIONS

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I. INTRODUCTION

Shortly after the discovery of gaseous detonations

by Berthelot and Vielle and by Mallard and Le Chatelier,

Chapman and Jouguet gave a theoretical description

of a one-dimensional detonation. Their theoretical

model was very successful as far as the prediction

of detonation velocities is concerned. The under-

standing of the stability of a detonation is not so

well developed.

The stability of detonations is of interest in two

respects.

1) Limits of detonability are the stability limits

   of selfsustained detonations. They are of great

   importance concerning the problems of safety.

2) The understanding of the stability of detonations

   is essential for the elucidation of the propagation

   mechanism of the detonation itself.

II. THE ONE-DIMENSIONAL MODEL OF A DETONATION

In the classical model a detonation is described as

a shock wave behind which a rapid chemical reaction

occurs after an induction time. Pressure, density, and

temperature of the fresh gas are increased

suddenly by the shock wave. During the induction

time these values remain constant. But after the

spontaneous onset of the exothermic chemical reaction

the temperature increases, why the pressure and the

density decrease to lower values. In this model a

stable detonation can be described by the laws

of conservation of mass, momentum, and energy. The

Chapman-Jouguet law states, that for a stable detonation

the Rayleigh-line has to touch the Hugoniot-curve

for the final products. From this, it can be deduced that

the velocity of a detonation is a minimum velocity. The

basic assumption of this one-dimensional model is

that a thermodynamic equilibrium exists both in front

of and behind the reaction zone. However, the effects

of transport phenomena, the influence of the tube

diameter, the length of the reaction zone and the details

of the chemical reaction were not considered. Detonation

velocities calculated by this model correlate with
those obtained experimentally. However there exists
sometimes a distinct discrepancy between calculated
and measured values for the density and pressure.

III. A DETONATION AS A THREE DIMENSIONAL PHENOMENON

Many experiments, using a variety of experimental
techniques, have been performed to obtain a more re-
alistic model of a detonation. Two ways are normally
used for the experimental investigation of a detonation:
the first is concerned with the macroscopic behaviour
of a detonation and the second with the microscopic
phenomena. The "microscopic" point of view is con-
cerned mainly with the interaction of shock waves.
Deep insight into the interaction of shock waves was
obtained by the method of soot traces and instantaneous
Schlieren photographs, especially by using Laser
techniques [2] . Despite the great number of experiments
and thorough theoretical calculations one is not yet
able to give a theory describing a detonation as a whole

Especially the problems concerning the initiation

of a detonation and the limits of detonability

have not yet been solved. It is obvious, however,

that a detonation is a three dimensional process

with a very com lex microscopic structure. The front

of a stable detonation is uneven and determined by

shock wave interactions, i.e. as the Mach triple con-

figuration, periodic movements of shock waves at the

front of the detonation are coupled with transverse

oscillations in the burnt gas as observed by the

macroscopic phenomenon of spin. Transverse oscillations

in the reaction zone seem to be responsible for the

transport of energy to the detonation front, and a

detonation is stable as long as these transverse

oscillations are compatible with the boundary conditions.

IV. THE LIMITS OF DETONABILITY

In a plot of detonation velocity versus fuel gas

concentration the limits of detonability are characterized

by a jump in the detonation velocity. These limits

give the region of the gas mixture in which a
stable detonation is possible. At the limits of
detonability one always observes the lowest possible
frequency of spin which can be calculated by a
simple formula derived by Manson and Fay [3] .

$$v = \frac{1.84}{\pi} \frac{a}{d} \quad \text{(circular tube cross section)}$$

Until now the influence of the tube diameter, the
addition of inert gases to the explosive gas mixture,
the initial pressure and temperature on the limits of
detonability have been investigated [8] .

The principal results shall be summarized: the fuel
gas concentration at the limits is a linear function
of the reciprocal tube diameter for a wide range of
tube diameters. The slope of this straight line is a
measure of the influence of the tube diameter on the
limits of detonability. The detonation velocity
measured near the limits is in good agreement with
the values calculated from the Chapman-Jouguet-theory
The addition of inert gases can lead to a detonation
regime which is either smaller or larger than without
an inert gas. This is mainly given by the ratio of

the of cific heats behind the shock front and by

the dilution of the reactive components of the

gas mixture. A decrease in the initial pressure

leads to a smaller detonation regime. As there

exists a critical diameter for the propagation

of a stable detonation, there also exists a critical

pressure. This is the lowest pressure for which a

stable detonation is possible.

V THE INFLUENCE OF INITIAL TEMPERATURE ON THE LIMITS

OF DETONABILITY

Until now little is known about the influence of

initial temperature on the limits of detonability,

but the influence of initial temperature on the

detonation velocity has been investigated several

times [4] . Dixon measured the detonation velocities

for mixtures of $C_2H_4 + 2O_2$ and $C_2N_2 + O_2$ at initial

temperatures of 10 and 100 ^{o}C and at 1 atm. initial

pressure. Laffitte investigated the detonation velocities

of $2H_2 + O_2$ (450 - 475 ^{o}C initial temperature) and

$CH_4 + 2O_2$ (525 - 530 ^{o}C initial temperature) at
normal pressure. Ragland, Cosens, and Cullen deter-
mined detonation velocities of stoichiometric hydrogen-
oxygen mixtures for low initial temperatures down
to the vicinity of the oxygen vapour saturation point at
initial pressures from 1 to 15 atm. Gordon and Zelesnik
calculated detonation parameters of hydrogen-oxygen
detonations from 0.01 to 100 atm initial pressure and
from 200 to 500 ^{o}K initial temperature. Another
investigation about the influence of initial temperature
on detonations in hydrogen-oxygen has been published
by Ladermann.

Summarizing all these investigations one can say:
at constant initial temperature the detonation
velocity increases with increasing initial pressure,
and at constant initial pressure the detonation ve-
locity decreases with increasing initial temperature.
The variation in the detonation velocities is about
1 to 2 % when the initial temperature is varied by
100 ^{o}K. It should be mentioned that this result is in

accordance with the Chapman-Jouguet-theory.

The main subject of this report is the influence
of initial temperature on the limits of detonability.
What can be expected from the Chapman-Jouguet theory ?
If Q is th heat of reaction per unit mass, $\varkappa$ the
ratio of the specific heats, D the detonation velocity,
a the velocity of sound, and index o refers to the fresh
gas, then the following relation holds approximately:

$$D^2 \approx 2 (\varkappa^2 - 1) Q$$

$$T_{CJ} \approx \frac{T_o \varkappa_o (\varkappa_o - 1) D^2}{(\varkappa_o + 1)^2 a_o^2}$$

In a first approximation the initial temperature T_o
has no influence upon D. a_o^2 is proportional to T_o
and therefore the temperature at the CJ-plane, T_{CJ},
is approximately independent of T_o. From this it can
be expected that a stable detonation should not be greatly
influenced by the initial temperature. But at the limits
of detonability the Chapman-Jouguet theory is not valid
and here a greater influence of the initial temperature

can be expected.

VI THE EXPERIMENTAL DESIGN

The influence of initial temperature on the limits

of detonability was investigated at 1 atm. initial

pressure and for initial temperatures of $T_o = 295\ ^oK$

(room temperature), $T_o = 195\ ^oK$ and $T_o = 135\ ^oK$.

The experiments were carried out for the following gaseous

mixtures: hydrogen - oxygen, hydrogen - air, and methane -

oxygen. For comparison with these initial temperatures,

the boiling points of the components of the gaseous

mixtures are given here:

Methane 111.7 ^oK

Oxygen   90.2 ^oK

Hydrogen 20.7 ^oK

Nitrogen 77.4 ^oK

VI 1) THE PHOTOGRAPHIC TECHNIQUE

The combustion processes in the tubes were recorded

by a rotating drum camera which is described in

detail in [5] . With the film moving perpendicular

direction of move-
ment of the film

Film

images of the
plexiglass tubes on
the film

direction of
propagation of
the detonation

luminous zones
at the time t_1

luminous zones
at the time t_2

path of the luminous
zone on the film

x

t
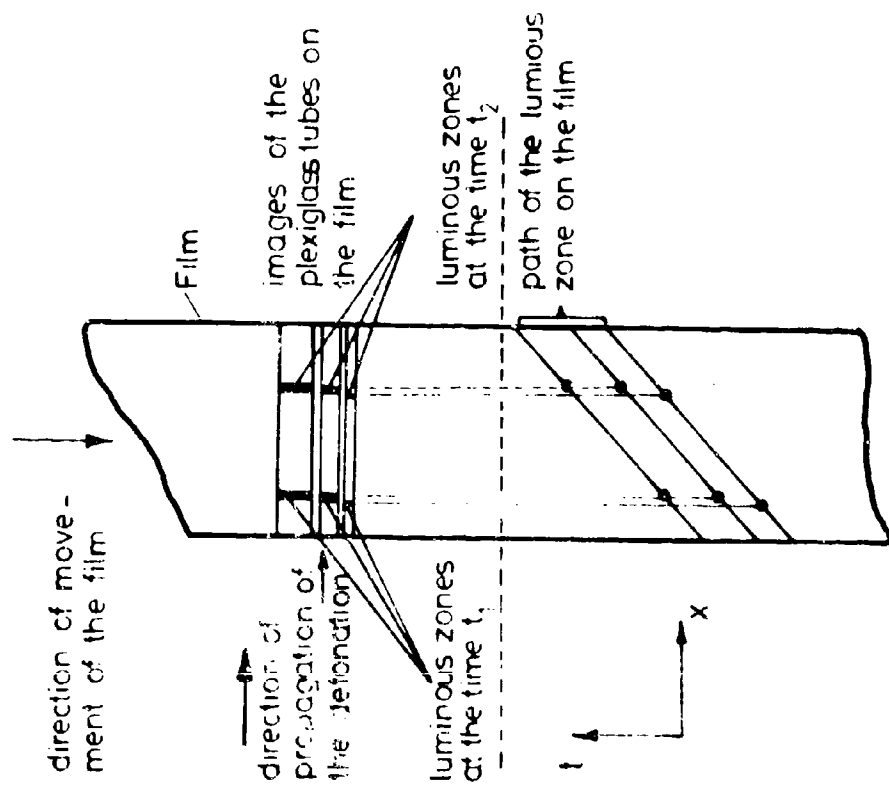
Fig. 1   Illustration of the formation of
a space - time $(x-t)$ diagram
on the film

to the image of *the* detonation front one obtains

a space-time-diagram of the luminous zones of the

combustion process. This luminous detonation front

gives an inclined path on the film, and the slope

of this path is dependent on the relative velocity

of the film and the image of the luminous front. This

is demonstrated schematically for a stable detonation

in Fig. 1. The detonation velocity can be calculated

from the slope of the path, the ratio of the original

tube length to its image on the film, and the frequency

of the rotating drum camera. This photographic in-

vestigation of the stability of a detonation has

some advantage to the registration of the detonation

velocity by an electric device. On the film not only

the variation of the detonation velocity at the limit

of detonability but also the structure of the com-

bustion process is recorded. Studying the structure

and the velocity of the detonation front one obtains a

very critical proof of stability. From this point of

view a detonation is a stable detonation if the
luminous path on the film is line and if its
structure is regular (Picture 1). It is also under tandable
that there might be a slight difference in the
results from this method to measurements made by the
registration of the electrical signals of probes.

VI 2) THE DETERMINATION OF THE FUEL GAS CONCENTRATION

To avoid explosions hazards the explosive gas
mixtures were not stored in tanks. The flowing gases
were premixed roughly by means of flowmeters and the
fuel-gas concentration was determined finally by a
Zeiss Haber-Loewe-Gasinterferometer, which was
thermostated at 24 $^{\circ}$C. This interferometer was cali-
brated for each measurement with oxygen as reference
gas. By this method the fuel gas concentration could
easily be determined to an accuracy of $\pm$ 0,5 %.
Commercial gases were used which were dried with a
mixture of dry ice and methyl alcohol.

Picture 1: Two self-sustained detonations
in H_2 - air, $T_0 = 295$ ^{O}K, $p_0= 1$ atm.

22, 1 Vol % H_2 d = 26 two marks

d = 16 one mark

VI 3) THE COOLING OF THE GASES

To cool down the explosive gas mixture it was

necessary to use inflammable refrigerants. In

the first step of the research the limits of

detonability of methane-oxygen mixtures were investigated

at T_o = 295 ^oK and 195 ^oK. This system is much easier

to investigate than the other ones because of the

higher luminosity of the detonation front. On the

other hand the detonation pressures are expected to

be higher than for $H_2 - O_2$ mixtures. It was decided

to use two experimental designs for cooling down the

gases. The first experimental arrangement was a

relative simple one and was used for T_o = 195 ^oK.

It was a 10 m long isolated container of aluminium

in which the detonation tubes were cooled down with

dry ice. For the heat insulation of the experiments

at T_o = 155 ^oK, a 7 m long cylindrical Dewar vessel

was constructed and liquid nitrogen was used as a

refrigerant. The Dewar was found to be more efficient

at the lowest temperature because of its better in-

sulating properties than that of the commercially

available materials. The thickness of the inner

Dewar wall was as thin as possible to avoid too

high a consumption of liquid nitrogen. This arrange-

ment was found to be most satisfactory at lowest

temperatures. For the experiments at $T_0 = 195$ ^0K the

detonation tubes were constructed out of metal to

which plexiglass tubes were connected.

Because at the lower initial temperature ($T_0 = 135$ ^0K) the

connections between the metal tubes and the plexiglass tube

became loose, it was decided therefore to use plexi-

glass - XT throughout. No influence of the material

of the tubes on the limits of detonability could be

detected at room temperature.

VI 4) THE INITIATION OF THE DETONATION

The problem of cooling down the tubes gives an upper

limit for the tube length. It was therefore decided

to initiate the detonation in the test mixture using

a stoichiometric mixture of the same components. However,

the tubes must have a minimum length due to the existence

gas mixture for ignition

spark plug

coil of wire

ignition section

0.60m

to the pump

1.30 m

cellophane diaphragm

starting chamber

83 mm

gas mixture under test

10 m

metal tubes

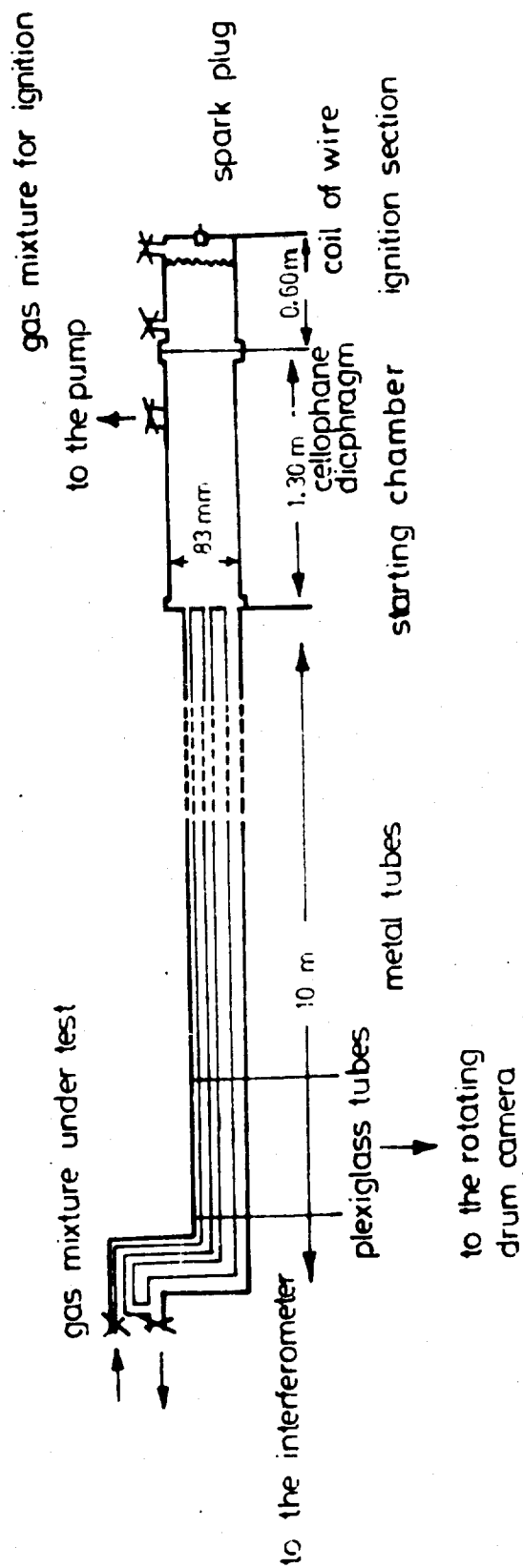plexiglass tubes

to the interferometer

to the rotating drum camera

Fig. 2   Sketch of the Apparatus for $T_0 = 195\,°K$

of overdriven detonations in the first part

of the tubes. For the $H_2 - O_2$ and $CH_4 - O_2$ systems

tubes of "m length and an inner diameter less than

30 mm were found to be adequate. The general scheme

of the experimental arrangement is illustrated in

Fig. 2. In order to reduce the length of the ignition

section a coil of wire was inserted [6] . This section

was seperated from the starting chamber by a dia-

phragm of cellophane or aluminium. In our case the

kind of diaphragm did not influence the measurements.

VI 5) THE EXPERIMENTAL DESIGN FOR $T_o = 195 \, ^oK$ AND THE
DESCRIPTION OF MEASUREMENT

A sketch of the apparatus is given in Fig. 2. Each

experiment was performed with three tubes of aluminium

of 4, 5, 0, or 10, 16, 26 mm inner diameter and

1 mm wall thickness. These tubes were situated one

above the other with a distance of 1 cm between them. To

the length of the tubes of about 10 m, several tubes

tube of plexiglass

PVC

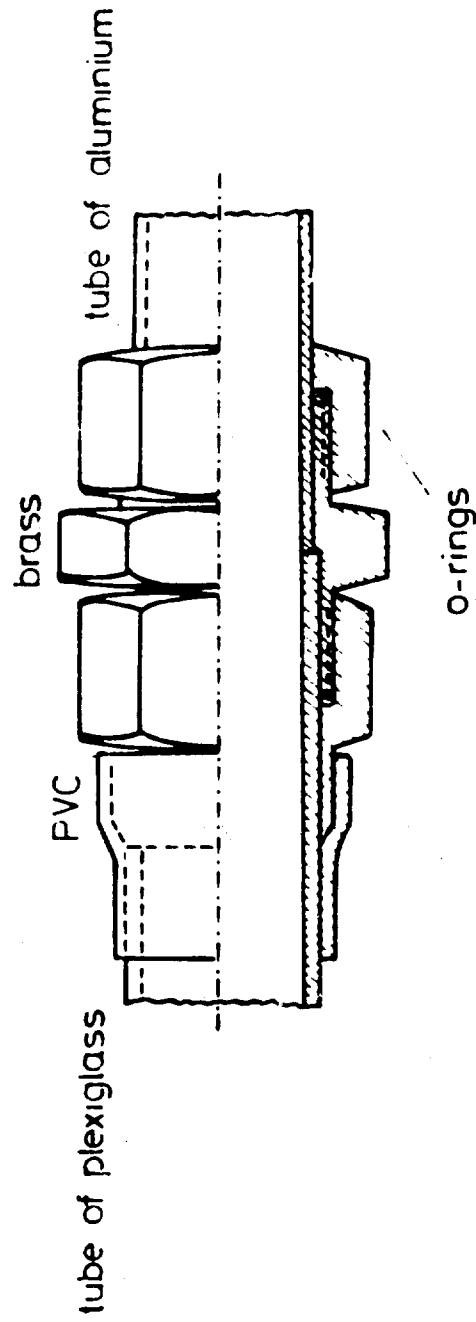brass

tube of aluminium

o-rings

Fig. 3   Sketch of a Stuffing Box

of aluminium had to be joined with stuffing boxes

and o-rings. The connection between the plexiglass tube and

the metal tube was made in a same manner (Fig.3). It was

taken care of the connections between two tubes to be smooth.

At the beginning of each experiment the tubes were put

into the isolated container of aluminium with a cross

section of 80 x 80 mm and 10 m length and the observation

window at one end, which was normally closed by the

isolating material (Exporit).

After the refrigerant of crushed dry ice was packed around

the detonation tubes, the container was closed by plates

of the isolating material. In this way the low temperature

inside could be maintained without significant losses. The

temperatures along the tubes were measured with thermo-

couples and monitored by a 12-point-recorder. After the tubes

were cooled down to the temperature of dry ice, the premixed

dried gases streamed through the smallest tube into the

initiation section. From here the gases flowed through the

two other tubes into a thermostated copper capillary which

led directly into the gas interferometer. From the
interferometer the gas passed through a long PVC tube
into the open air. The gas flow through the inter-
ferometer was about 100 cm^3/sec. If the reading re-
mained constant for half an hour, the gas stream was
stopped and the valves were closed. The composition
of the mixture was then determined with the gas at
re t. During the time while the gas in the tube was
been cooled down to 195 ^oK, the initiation section
was filled. Prior to an experiment the isolating
material was taken out of the window of the container
and the outside of the plexiglass tubes was defrosted
by methyl alcohol p.A. The gas mixture in the initiation
section was ignited by a spark plug and the detonations
in the tubes were recorded by the rotating drum
camera.

After each measurement the tubes had to be warmed up to
room temperature and replaced if damage had been occured.

TOTAL VIEW

observation window

rubber plate

metal bellow

rubber plate

starting chamber

metal bellow

plexiglass tubes

ignition section

diffusion pump

rotary pump

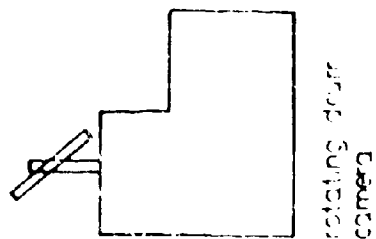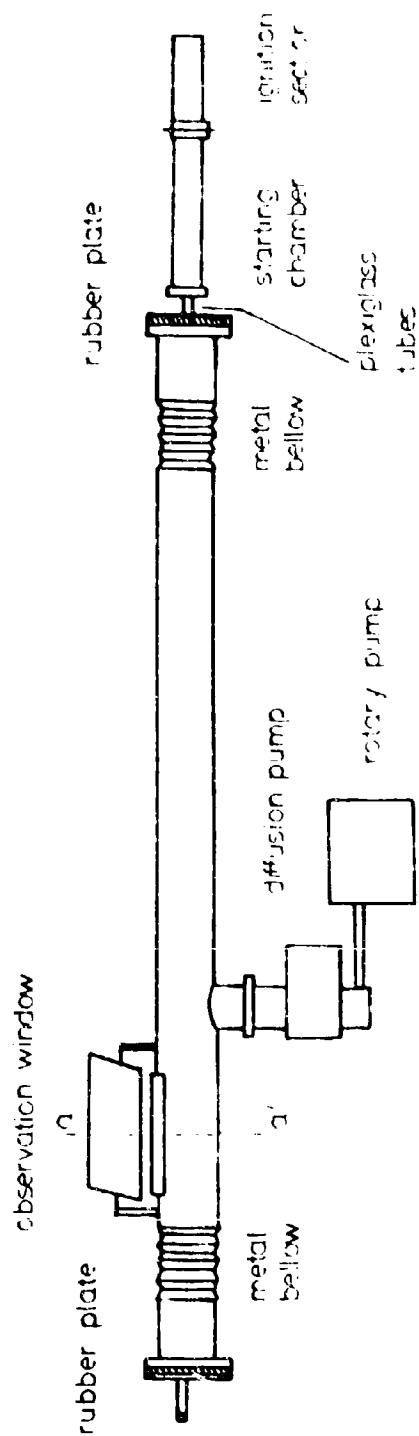section a - a'

rotating drum camera

Fig. 4   Sketch of the Apparatus

The water inside was then removed by pumping. The dried

tubes were then put back into the cooling system and

a new experiment could be started.

It is interesting that at the low temperature at the

limits of detonability dents were often observed along

the first   two or three meters of the aluminium tubes

behind the starting chamber. There is no doubt, that this

is an indication for the onset of detonations.

VI 6) <u>THE EXPERIMENTAL DESIGN FOR $T_o = 135\ ^oK$ (Fig. 4,4b)</u>

For the experiments at this temperature plexiglass XT

tubes of 7 m length with an inner diameter of 6, 10, 16,

and 26 mm and a wall thickness of about 2 mm were used.

To avoid too high a consumption of the refrigerant

a 6.50 m long cylindrical Dewar vessel with a window

of 60 cm length near one of its ends was constructed and

built in our work shop. This Dewar was made of brass with

an outer tube of 150 x 2.5 mm and an inner tube of 93 x

1.5 mm. The end plates of the Dewar vessel bet-
ween the outer and the inner tube, and the frame
of the window were made of German silver. The
pressure in the Dewar vessel was kept at 10^{-4} Torr
with a rotary oil pump and an oil diffusion pump.
For a Dewar vessel of this length the thermal
dilatation must be compensated for. The inner part
of the Dewar vessel was at 135 ^{o}K whereas the outer
part was at room temperature and both parts were con-
nected at the ends by German silver with 2 mm thick-
ness. For this reason flexible metal bellows (Tombak)
were soldered near both ends into the outer tube
of the Dewar vessel. The window was built in a manner
that it could overcome the resulting mechanical strains.
Liquid nitrogen and CF_2Cl_2(F12) were used as refrigerants.
The latter has a melting point at
- 155 ^{o}C with a comparatively high heat of fusion.
Thus an average temperature of - 140 $\pm$ 5 ^{o}C along the

straight tubes inside the Dewar vessel was a-
chieved. At 10^{-4} Torr in the Dewar vessel heat
losses by conduction were kept to a minimum and those
by radiation were reduced by polishing the inner
surfaces of the Dewar vessel before mounting.

VI 7) <u>DESCRIPTION OF MEASUREMENT AT 135 ^{o}K</u>

In one experiment we only used two tubes lying side
by side, which were filled with the gas under in-
vestigation in the manner described above. When the
gas stream was stopped, both the valves were closed
and the open ends of the plexiglass tubes were closed,
too. The tubes were then inserted into the Dewar
vessel which was already at the low temperature. The
ends of the Dewar vessel were closed by placing rubber
plates over them. These plates contained holes to
enable the detonation tubes to pass through them
(see Fig. 4). For cooling down of the tubes, a
sufficient quantity of the refrigerant was then filled
into the Dewar vessel. The pressure inside the closed
detonation tubes was reduced at the lower temperatures.

In order to overcome this decrease of pressure,
a flexible tube of PVC with a volume of approxi-
mately 10 l (inner diameter 16 cm) which acted
as a gasometer was connected to the detonation
tubes and filled with the same gas mixture. Thus
the initial pressure in the system could always
be maintained at 1 atm. by reducing the volume of
this gasometer. In contrast to the apparatus des-
cribed under 5) the two tubes for each experiment
were situated side by side. A surface mirror lo-
cated in front of the observation window was used
to deflect the light of the detonations to the ro-
tating drum camera.

After the cooling down of the tubes and the filling
of the initiation section with the stoichiometric
gas mixture, the insulating material in the obser-
vation window was removed and replaced by a plate
of plexiglass to prevent heat convection and ice
forming on the outside of the detonation tubes. The

detonations were then initiated and the subsequent

combustion processes were recorded photographically.

In both cases 5) and 7), the frequency of the rotating

drum camera was measured by an oscilloscope, using

Lissajous figures with a frequency generator as a

standard.

After each measurement the tubes were taken out of

the Dewar vessel, heated up to room temperature and the

water inside was removed by pumping. It is to be

mentioned that, at the limits of detonability, the

plexiglass tubes were generally destroyed within a

few meters after the starting chamber. This again was

the point where the detonation was started.

VI 8) THE PHOTOGRAPHIC MATERIAL

It is known that for the gas systems under investigation

the luminosity of the combustion processes near the

limits of detonability is very weak. This is especially

true for detonations in hydrogen - oxygen or hydrogen - air

mixtures were in the present investigation

a lot of trouble was experienced in obtaining

suitable photographs at the limits of detonability.

Therefore extremely high speed panchromatic films i.

e. Kodak 2475 Recording Film with DK 50 as a de-

veloper and Kodak High Speed Recording Film

with MX 642-1 as a developer were used. The develop-

ment was usually made at elevated temperatures.

For detonations with higher luminosity Kodak TRI-X

Ian with Kodak HC-110 developer was used.

VI 9) THE EVALUATION OF THE FILMS

Depending of which experimental arrangement was

used either two or three combustion processes were

recorded (see Picture 1). If the detonation is

a stable one, the path of the luminous detonation

front has a very regular structure and can be ap-

proximated by a straight line. When the composition

of the gaseous mixture approaches the value at the

limit of detonability, the image of the detonation

Picture 2: A spinning detonation in H_2 - air
60,8 Vol H_2, T_0 295 °K, p_0 1atm, d=26mm
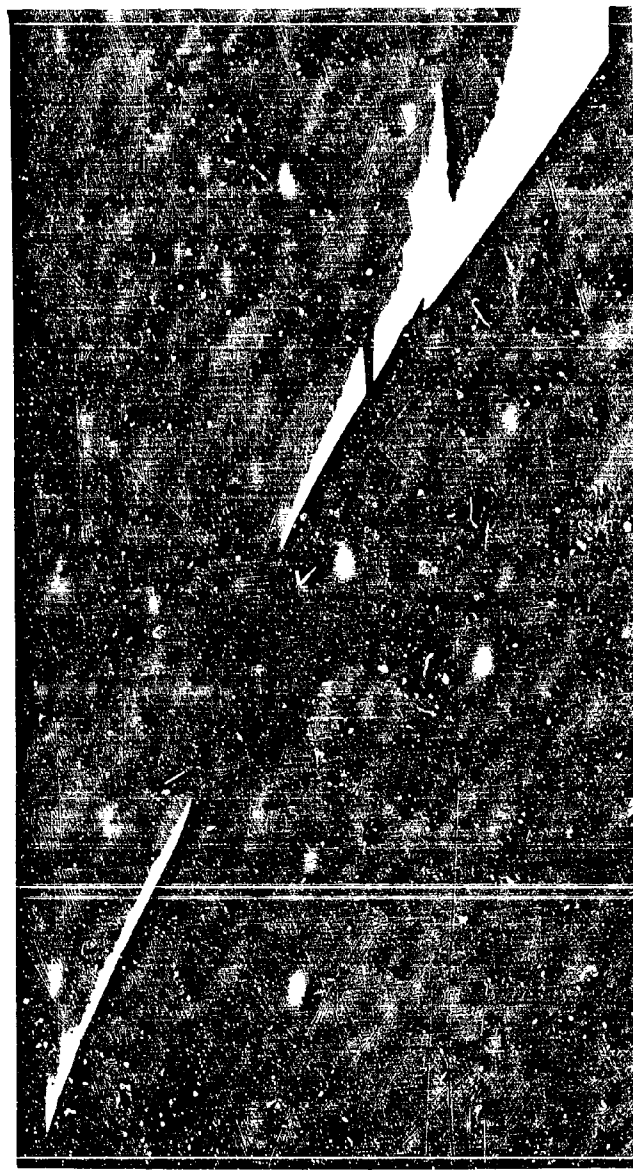
Picture 3: The onset of a detonation in $CH_4 - O_2$

9,7 Vol % CH_4, $T_o = 295\ ^oK$, $p_o = 1$ atm, d=10 mm

front gradually changes into a sinusoidal form
(see Picture 2). Beyond the limits of detona-
bility one observes detonations dying off or
starting with an alternating velocity of pro-
pagation. Finally only accelerated flames are
recorded (Picture 3), which are much slower than
detonations and which do not have such a marked
structure, until nothing is to be seen on the film.
Especially at the limits of detonability of hydrogen -
oxygen mixtures a fairly wide range of detonative
combustion beyond the limits of detonability was
observed. This is a very important fact for safety
purposes. Generally speaking as long as short sections
are considered there is only a gradual change from
the regime of a stable detonation to the regime
of flames.

For the determination of the limits of detonability
as a function of the tube diameter, the fuel gas
concentration was first chosen such, that in at least
one of the two or three tubes a stable detonation

could be expected. The limits of detonability

for one tube diameter were determined by averaging

between the two points having similar values of

the fuel gas concentration, of which one belonged

to a stable and the other to an instable detonation.

VII EXIERIMENTAL RESULTS

VII 1. DETONATION VELOCITY

The detonation velocity did not depend very strongly

on the initial temperature. This is in good accordance

with measurements discussed above. With an accuracy

of measurement of about $\pm$ 2 % we generally did not

find any influence of the tube diameter on the deto-

nation velocity. Only here and there just at the limits

of detonability a very slight dependence of the tube

diameter on the velocity of stable detonations was

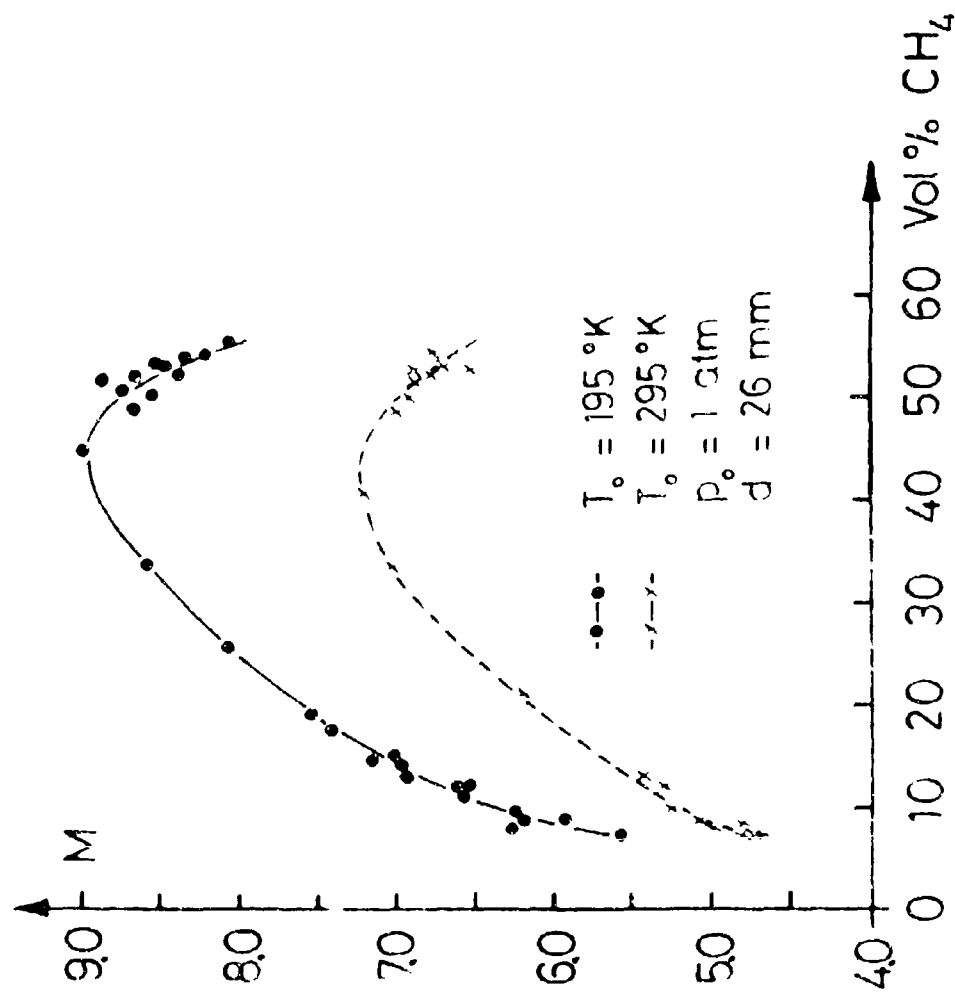found. In Fig. 5 and 6 the Mach numbers for detonations
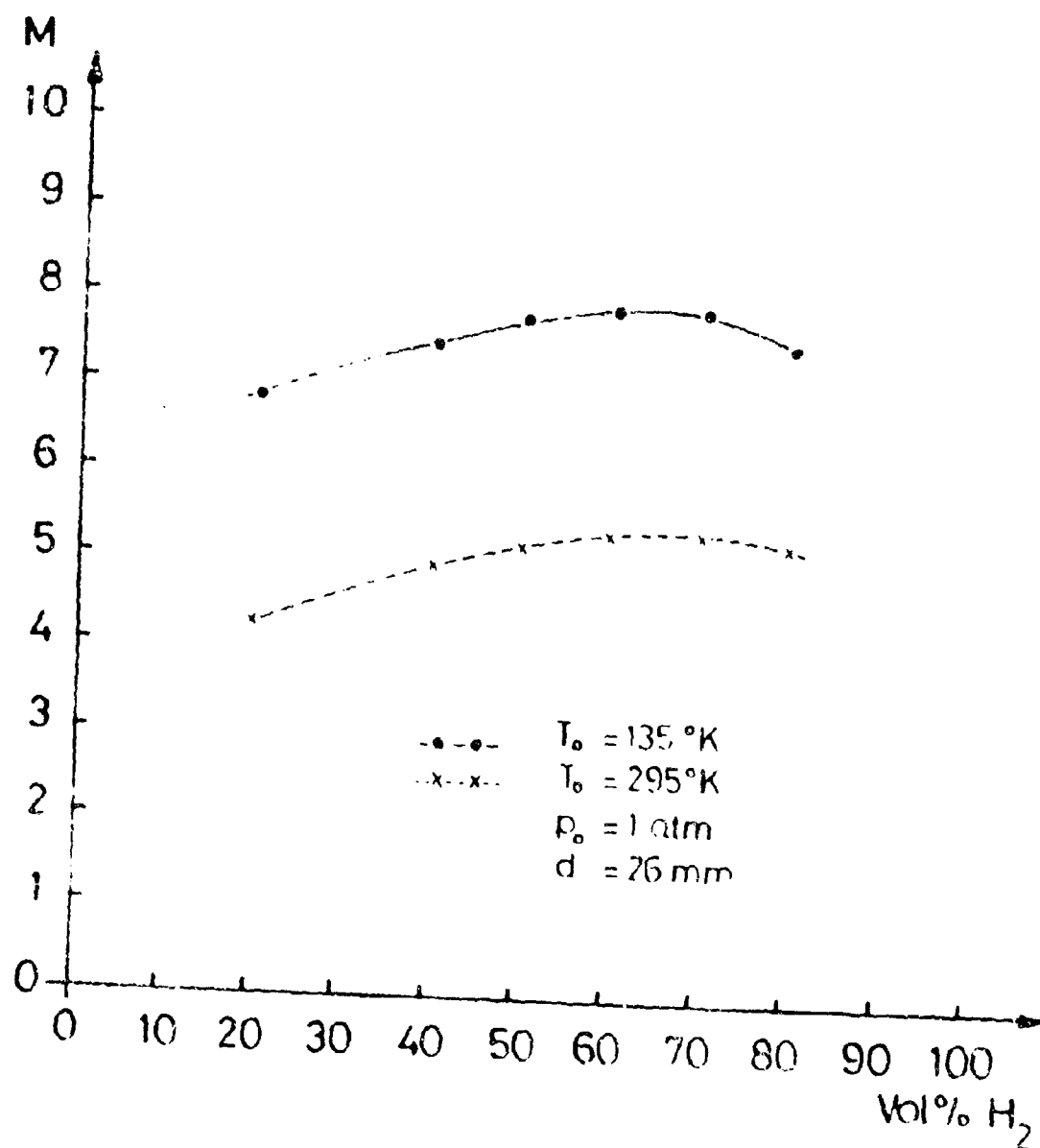
Fig. 5   Mach Number $M = D/a_o$
$M = f(C_{CH_4})$

Fig. 6   Mach Number $M = D/a_o$
          $M = f(C_{H_2})$

in $CH_4 - O_2$ and $H_2 - O_2$ mixtures are plotted

against fuel gas concentrations, with the initial

temperature as a parameter. These Mach numbers

determine the strength of the front shock waves.

The Mach number is defined as the ratio of the

detonation velocity and the velocity of sound in

the unburnt gas.

VII 2. <u>TEMPERATURE BEHIND THE SHOCK WAVE</u>

With these measured detonation velocities the

temperature behind the front shock wave was calculated

for $CH_4 - O_2$ (Fig. 7), assuming that just behind the

shock wave there is no chemical reaction. The

temperature behind the shock wave is nearly inde-

pendent of the initial temperature. This temperature

corresponds to the temperature at which the chemical

reaction in the detonation zone first starts, assuming

that the one-dimensional treatment of a detonation

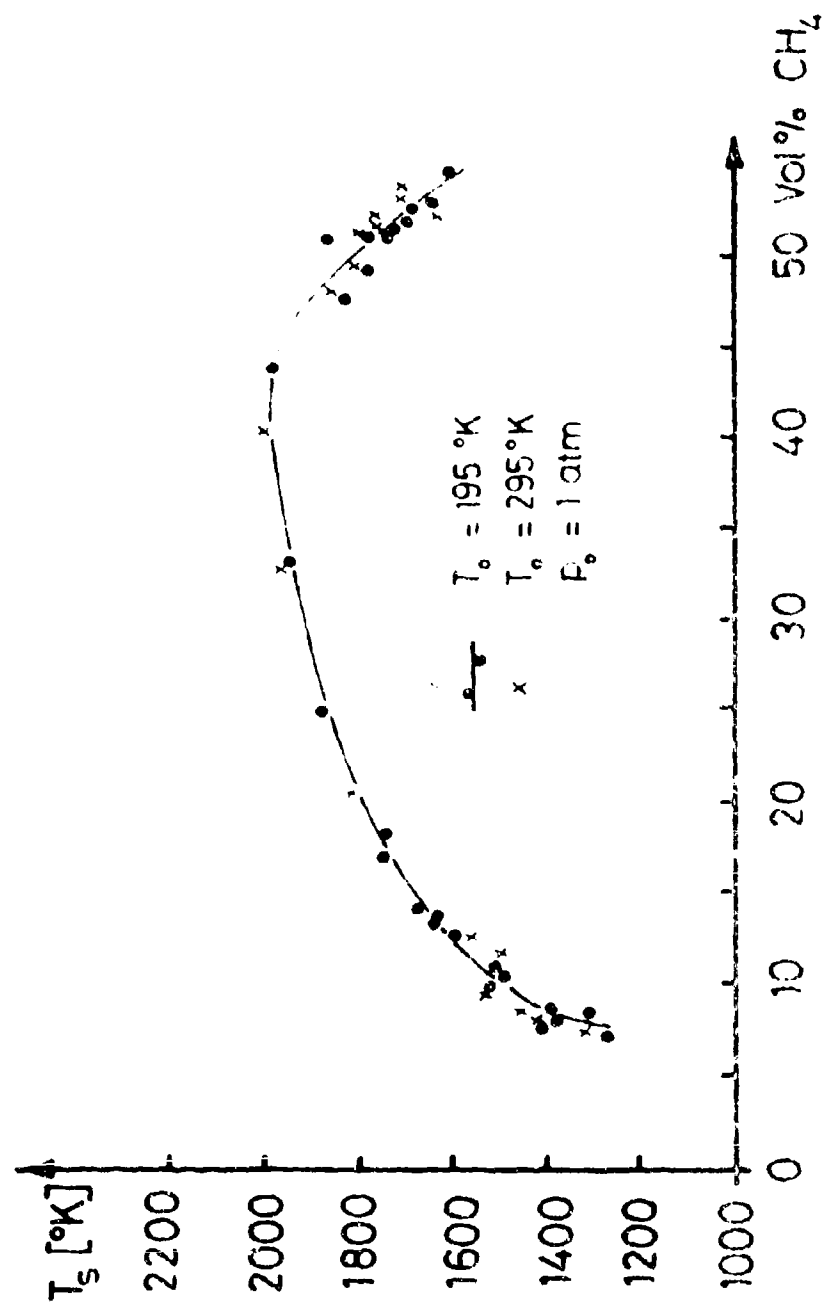is valid. On the other hand the pressure behind the

Fig. 7 Temperature behind the Front Shock Wave

shock wave is very strongly influenced by the
initial temperature.

VII 3. THE LIMITS OF DETONABILITY

The limits of detonability are represented in
terms of fuel gas concentration as a function
of the reciprocal tube diameter with the initial
temperature as a parameter (Fig. 8, 9, 10). In these
plots the upper and the lower limits of detonability
are represented by straight lines, the slopes of
which are a direct measure of the influence of the
tube diameter. For all these systems under in-
vestigation the concentration regime of stable
detonations becomes narrower for the lower initial
temperatures. Also the influence of the tube diameter
on the limits of detonability is a little more
pronounced at the lower initial temperature. The
measured values of the fuel gas concentrations at
the limits of detonability are tabulated in Table
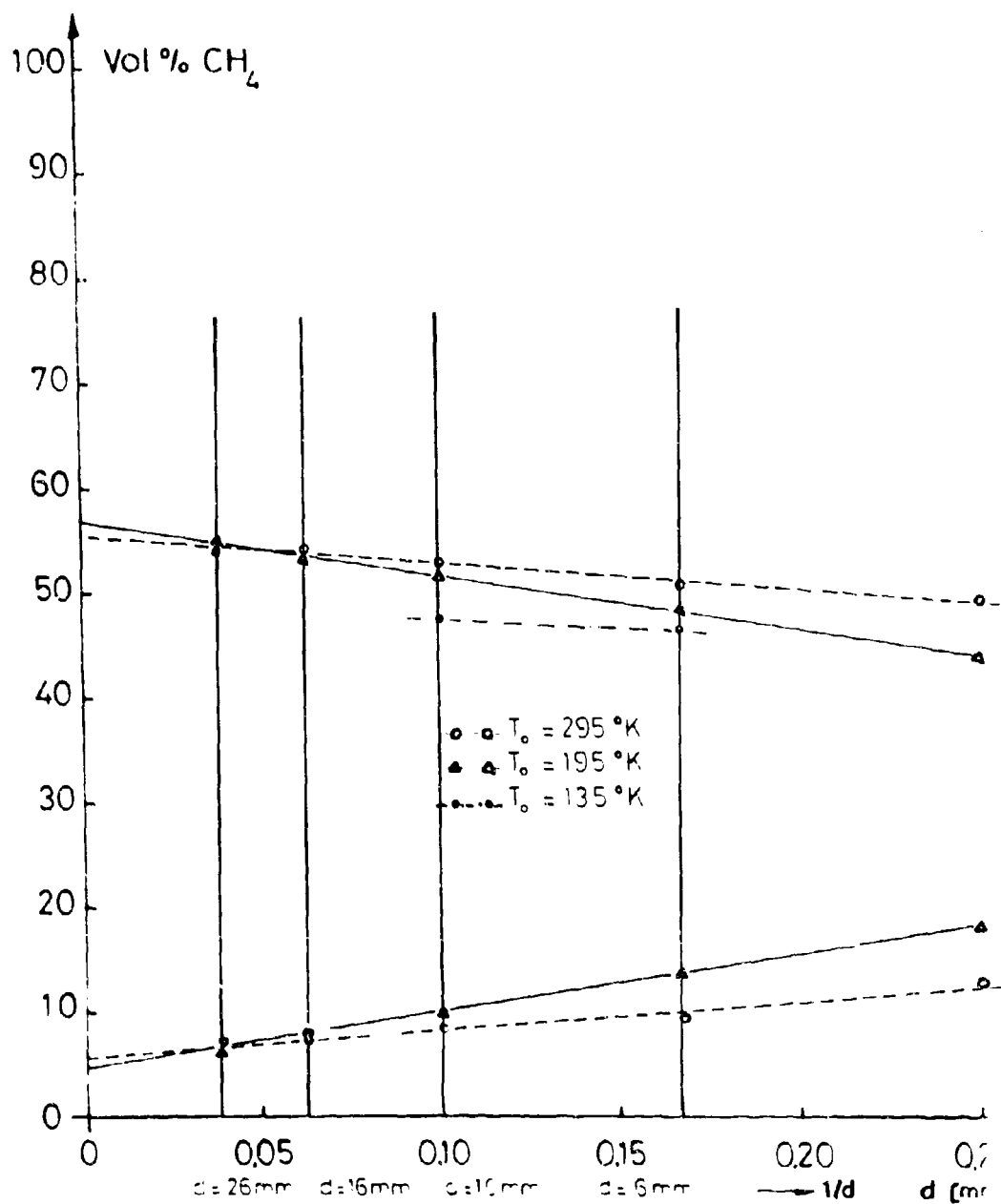1. It must be pointed out that the values for

Fig. 8   Limits of Detonability of $CH_4 - O_2$

Fig. 9 Limits of Detonability of H_2-O_2
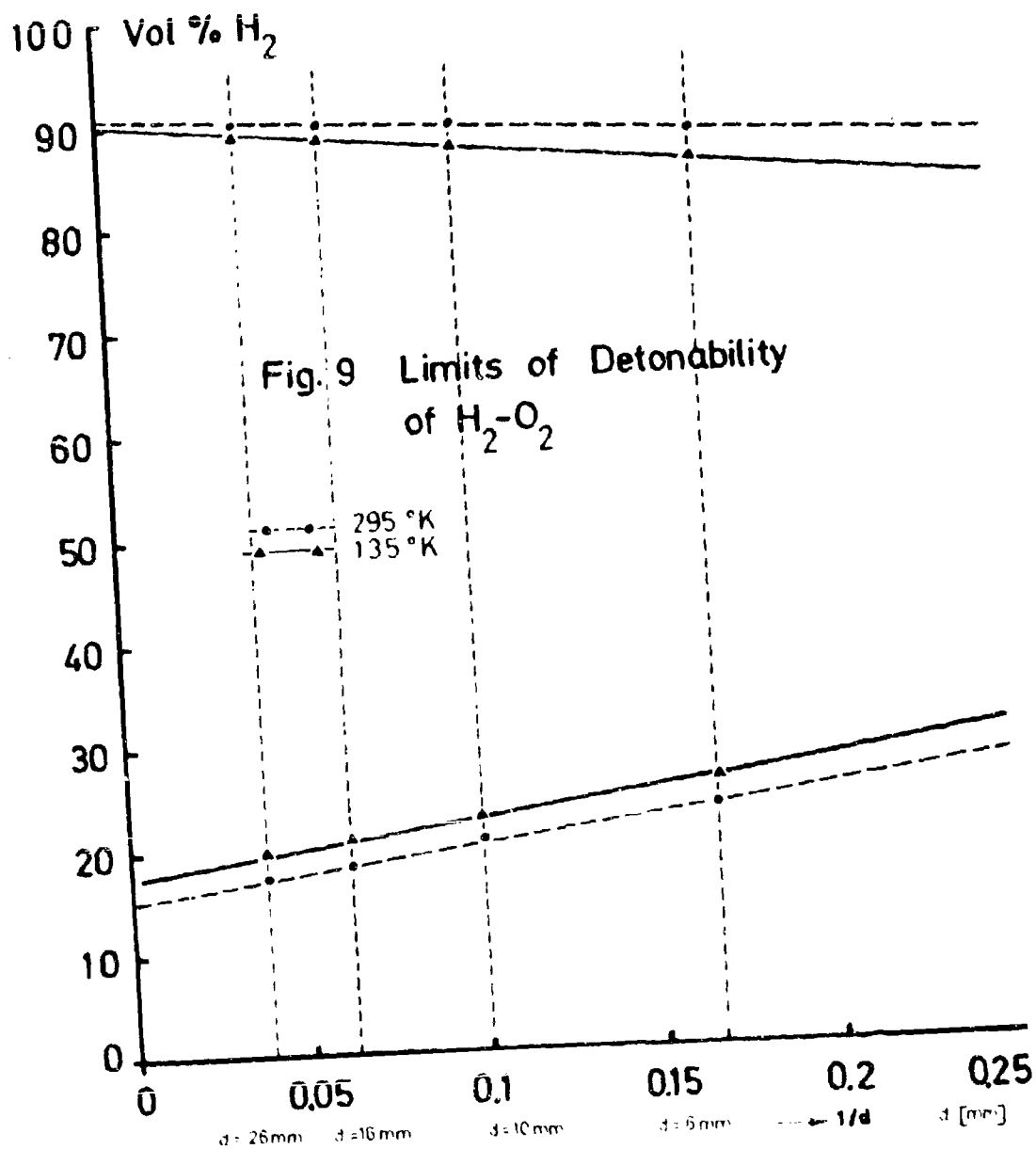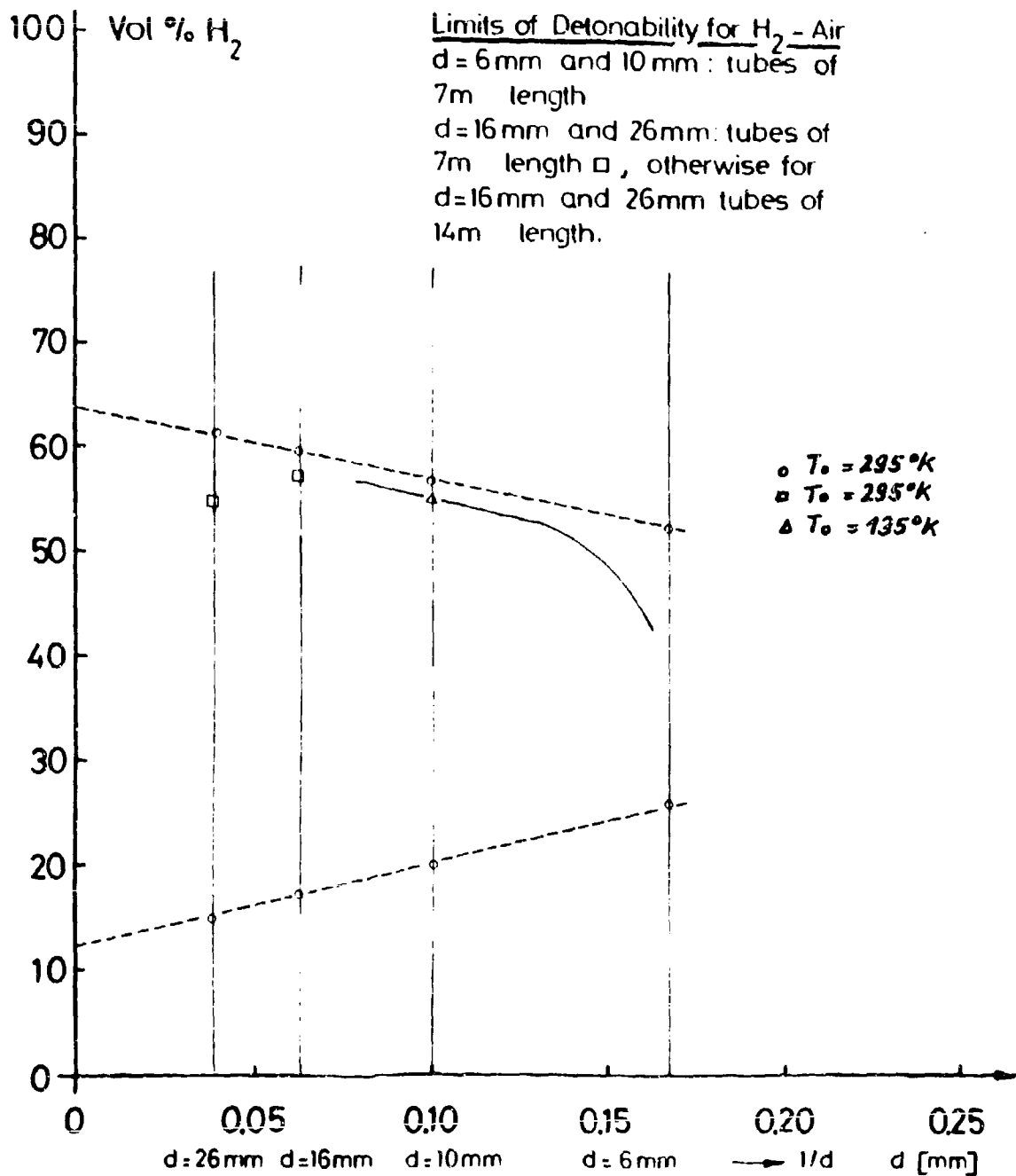
Fig. 10   Limits of Detonability for H_2 - Air

Table 1   The measured limits of detonability

$CH_4 - O_2$

T_o [°K]	d [mm]	Vol % CH_4	
		upper limit	lower limit
295	4	48.7	12.2
	6	50.7	10.0
	10	52.7	8.3
	16	53.6	7.3
	26	54.3	6.7
	∞	55.3	5.6
195	4	43.7	18.3
	6	48.0	13.7
	10	51.5	10.0
	16	53.4	8.0
	26	54.7	6.7
	∞	56.7	4.7
135	6	46.2	-
	10	47.3	-

$H_2 - O_2$

T_o [°K]	d [mm]	Vol % H_2	
		upper limit	lower limit
295	6	87.9	23.3
	10	89.1	20.0
	16	89.7	18.3
	26	90.0	17.0
	∞	90.7	15.3
13	6	85.0	26.0
	10	87.3	22.7
	16	88.4	20.7
	26	89.0	19.6
	∞	90.4	17.5

Table 1

H_2 - air

$T_0 [^{o}K]$	length of the tube	d $[mm]$	Vol.% H_2	
			upper limit	lower limit
295	7 m	6	52.6	25.8
	7 m	10	57.0	20.3
	14 m	16	59.4	17.3
	14 m	26	61.0	15.3
	7 m	16	57.3	-
	7 m	26	54.5	..
		∞	63.6	12.4
135	7 m	10	55.1	-
	7 m	6	did not detonate	

$d \longrightarrow \infty$ or $1/d \longrightarrow 0$ are extrapolated
values and that they have no real physical
significance. This can be seen very easily
by the fact that detonations in tubes with
very large inner diameters must be spherical
detonations and these cannot be compared to
detonations in tubes of small diameters.

For the system methane - oxygen only the upper limit
of detonability for $T_o = 135 \, ^{\circ}K$ was measured for
tubes of 13 and 6 mm inner diameter. This was done
because the impact of the detonations was too high
so that the Dewar vessel would have been damaged
by more measurements.

The system hydrogen - air is a very interesting
one. At room temperature the length of the tubes (7m)
was insufficient for $d = 16$ and 26 mm as   can be
seen in the plot. For this reason only the upper
limit of detonability was investigated at $T_o = 135 \, ^{\circ}K$

for tube diameters of 6 and 10 mm. For d = 6 mm

and T_0 = 135 ^{O}K the gas mixture did not detonate.

To demonstrate the influence of initial temperature

on a stable detonation some values just behind the shock

front have been calculated for $CH_4 - O_2$ at the limits

of detonability (Table 2).

VII 4. <u>THE CRITICAL TUBE DIAMETER</u>

For tubes with an inner diameter less than the critical

diameter a stable detonation is not possible. It is

very difficult to measure this diameter directly,

but from our measurements it can be estimated from

Fig. 11, 12, 13. Thus the influence of the initial

temperature on the critical tube diameter is ob-

tained, and this will provide new informations for

safety problems. The estimated critical tube diameters

are shown in Table 3.

Table 2: Calculated values behind the front shock wave (the detonation velocity is measured)

$p_0 = 1$ atm

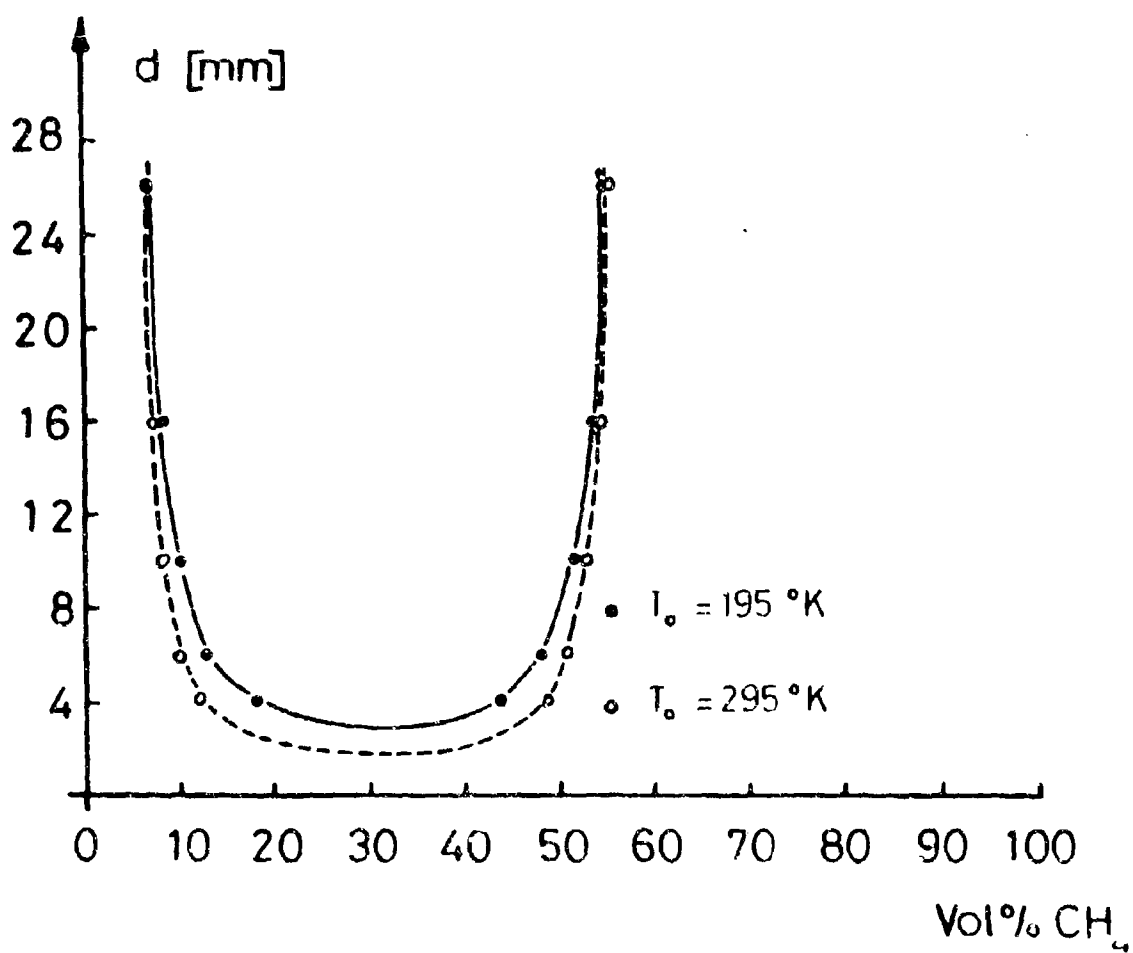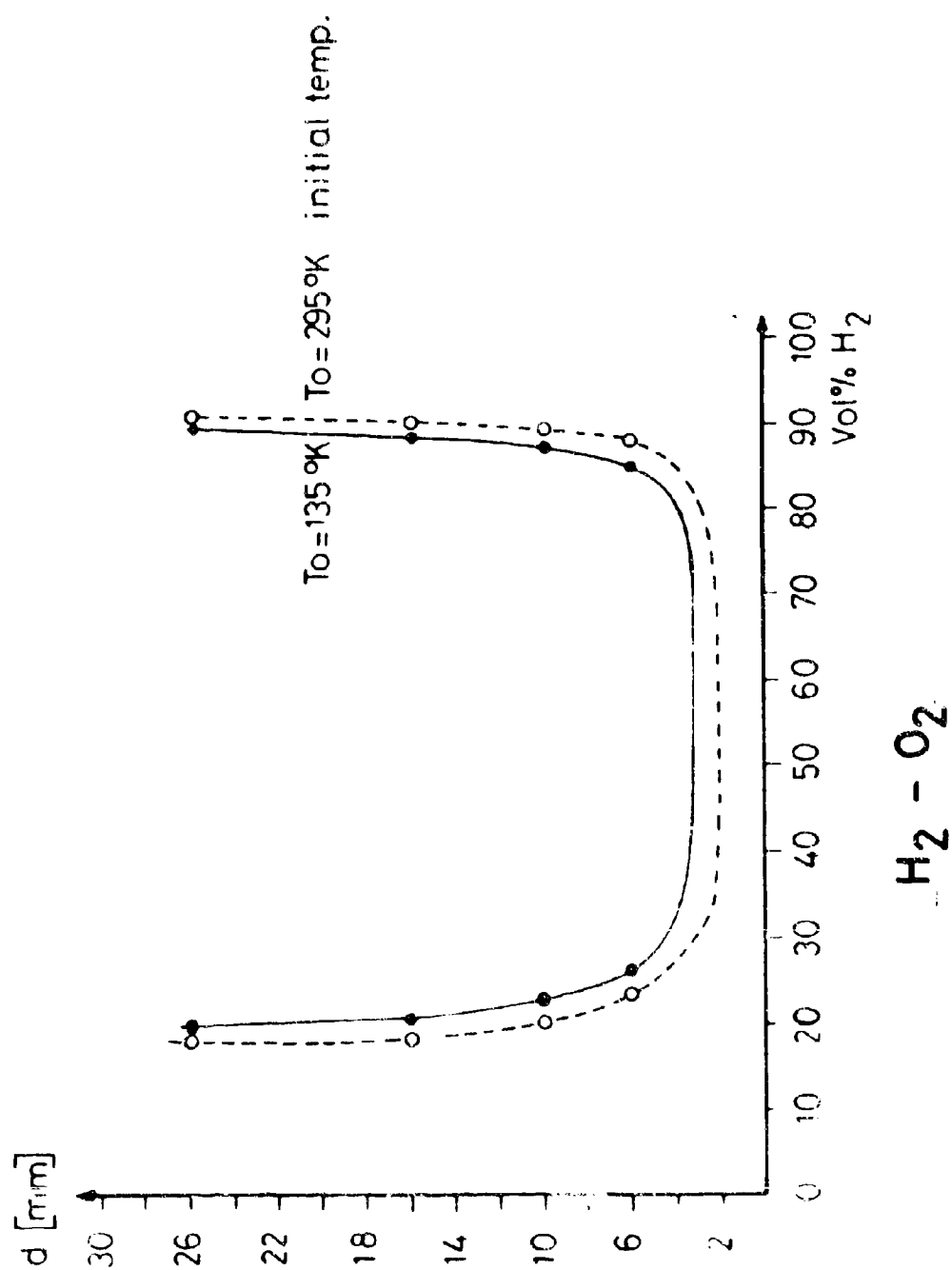| $T_0[°K]$	Vol.% CH_4	d[mm]	D[km/sec]	$M=D/a_0$	P_{st}/P_0	$T_{st}[°K]$	T_{st}/T_0	ρ_{st}/ρ_0	$p_{st}[10^{-3} \text{ g cm}^{-3}]$
Lower Limit:									
195	18.4	4	2.09	7.60	69.1	1760	9.0	7.7	14.2
195	13.4	6	1.92	6.94	60.7	1630	8.4	7.3	13.6
195	10.1	10	1.78	6.47	51.2	1490	7.7	6.6	11.6
295	12.2	4	1.84	5.47	34.8	1620	5.5	6.3	7.8
295	9.0	6	1.70	5.10	31.5	1470	5.0	6.3	8.0
295	7.2	10	1.55	4.80	26.1	1300	4.4	5.9	7.5
Upper Limit:									
195	49.7	4	2.66	9.00	109.3	1990	10.2	10.7	16.7
195	48.0	6	2.63	8.87	94.2	1900	9.7	9.7	14.7
195	51.3	10	2.60	8.60	90.4	1760	9.0	10.0	14.9
295	43.7	4	2.65	7.00	65.4	1380	6.4	9.9	9.9
295	51.0	6	2.55	6.91	59.1	1770	6.0	9.7	9.9
295	52.7	10	2.47	6.67	53.4	1690	5.7	9.3	9.1

Fig. 11 $CH_4 - O_2$

Fig. 12
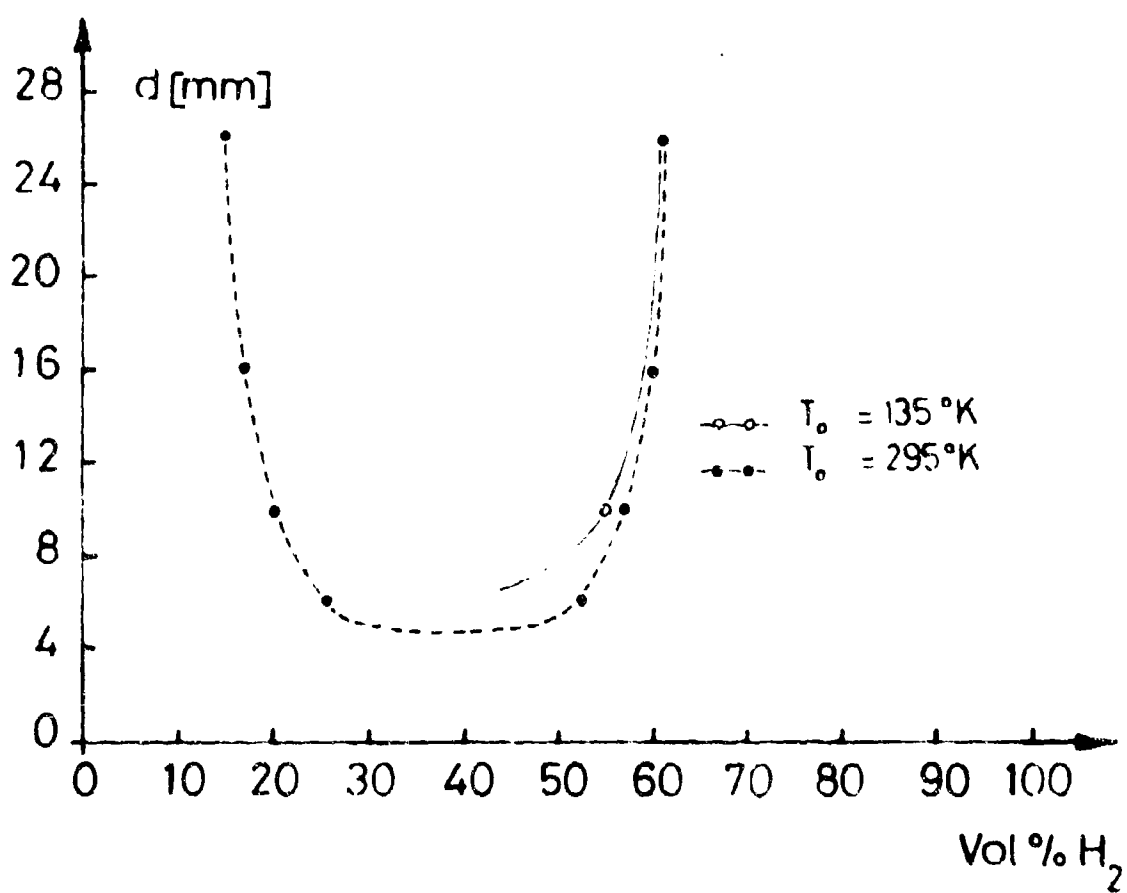
Fig. 13   H₂ – air

Table 3: The extrapolated critical tube diameter
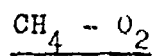
$\underline{CH_4 - O_2}$

$T_0 = 295 \, ^cK$ $d_{cr} \approx 2$ mm

     $195 \, ^cK$ $d_{cr} \approx 3$ mm

$\underline{H_2 - O_2}$

$T_0 = 295 \, ^cK$ $d_{cr} \approx 2$ mm

     $135 \, ^cK$ $d_{cr} \approx 3$ mm

$\underline{H_2 - air}$

$T_0 = 295 \, ^oK$ $d_{cr} \approx 6$ mm
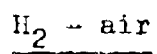
$T_0 = 135 \, ^oK$ $d_{cr} \approx 5$ mm

VIII DISCUSSION

The classical hydrodynamic theory of a stable

detonation is able to make good predictions

concerning detonation velocities, pressures, and

densities. The calculations for various initial

temperatures give results which are in good

agreement with the experimental values. In the

case of a stable detonation the phenomenon of

multiheaded spin occurs if the gas mixture is not

too close to the limits of detonability. In this

case the model of a one-dimensional shock wave, followed

by a rapid chemical reaction, is a good approximation

and it seems reasonable to consider a Chapman-Jouguet

plane. This theory does not say anything about the

limits of detonability. The existence of these

limits can be   attributed to phenomena which are not

considered in this classical theory. These phenomena

include reaction kinetics, hydrodynamic perturbations,

and the coupling of transversal shock waves with

the possible acoustic oscillations in the burnt

gases giv?` by the boundary conditions. From the point

of view of the reaction kinetics one has to consider the

heat release by the exothermic chemical reaction and the

time for the heat release, because this is the

energy source for the front shock wave. If these waves

are to influence the stability of a detonation, the

interaction of waves must be   considered between the

shock wave at the front and the place where the local

Mach number reaches the value one. This follows from

the Chapman-Jouguet theory. All these phenomena

cannot be considered separately, and this is the cause

for the complex nature of a detonation and the diffi--

culty involved in explaining the existence of the limits

of detonability. At the limits of detonability

one always observes single headed spin. The detonation

front is given by a shock wave configuration like

the Mach triple configuration. Furthermore there is

a measurable local separation of the shock wave at the

front of the reaction zone. The fact, that at

the limits of detonability the ratio of the wave

length of the acoustic oscillation in the burnt

gases to the tube diameter always reaches a critical

value, indicates a coupling of the shock waves at the

front with these acoustic oscillations.

From these facts one can try to explain the influence

of initial temperature on the limits of detonability,

Why do these limits become narrower for lower initial

temperature ? We have seen that the temperature be-

hind the shock wave at the detonation front is almost

independent of initial temperature, and this temperature

is determinative for the chemical reaction. From this

point of   view the reaction could only be influenced

by the higher pressure behind the shock wave at the

lower initial temperature. Because the reaction time

is proportional to p^{-n} this might prove to have a

slight influence. But then the detonation limits

would become wider with lower initial temperature .

There must be a dominant effect into the other
direction. Investigations of detonation spin
with the film moving in the direction of the
detonation front with the same velocity demonstrated,
that also at low temperatures there is a periodically
moving luminous zone in the detonation front. This
luminous zone is the zone of strong chemical reaction.
This picture of an oscillating zone of strong chemical
reaction in the detonation front is supported by the
reaction zone behind a triple Mach configuration
as demonstrated by Edwards a.o. [7] . At the limits
of detonability one triple Mach configuration goes
to and fro between the walls of the tube, and the
zone of the strongest chemical reaction also oscillates.
If the reaction zone is near the wall of the tube,
it might be influenced by the cool boundary layer,
and for this reason the lower initial temperature
might lead to a narrower detonation regime. Another
possibility as to why the lowering of the initial

temperature reduces the stability of a detonation is
the existence of rarefaction waves coming from the
cold wall into the zone between the front shock waves
and the regime of Mach number one. This should be
possible at the limits of detonability because of the
greater separation of the front shock waves and the
Chapman-Jouguet plane. Furthermore the damping of the
acoustic waves in the burnt gases must also be con-
sidered.

Only the influence of low initial temperatures on the
limits of detonability has been investigated. What will
happen if the initial temperature is increased ? There
is one criti al case. If the initial temperature
reaches the value for the self-ignition of the
detonative gas mixture, no detonation can be generated
anymore. In a similar manner, the onset of a detonation
from a laminar burning flame is restrained by higher
initial temperatures. Thus the detonation regime might

be expected to become narrower for higher

initial temperatures. Then there should be

a range of temperature at which the detonation regime

reaches a maximum.

IX A METHOD TO APPROXIMATE THE LIMITS OF DETONABILITY

For a stable detonation the detonation velocity

is independent of the length of the tube and there-

fore independent of the kind of ignition, provided

that the ignition was strong enough to ignite the

gases. Therefore one should expect, that the limits

of detonability in very long tubes are independent

of the manner of igniting either by an initial detonation

or by a weak flame. To test this, we took a tube of

plexiglass of about 3 m length with one end closed.

Near the closed end the detonative gas mixture was

ignited by a pilot flame through a small hole in the

wall of the tube. By this weak ignition no shock waves

were produced at the point of ignition. The induction

time and the induction distance were measured from the

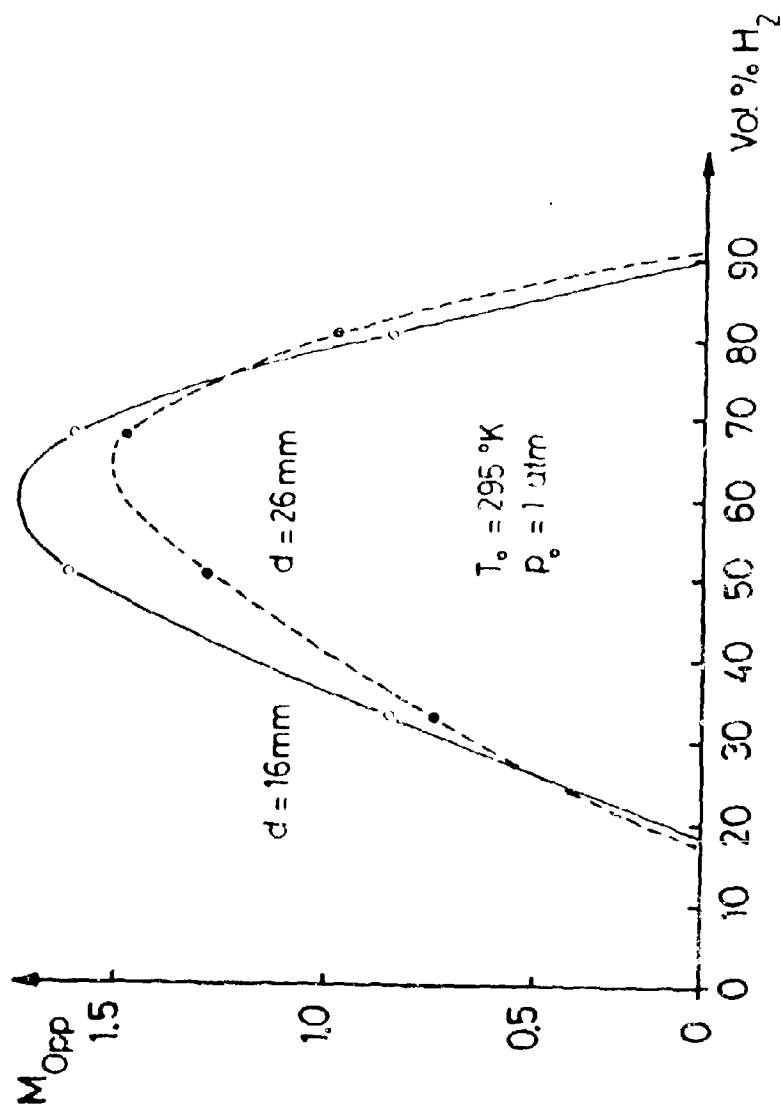point of ignition to the point of the onset of the

Fig. 14 $M_{Opp} = f(C_{H_2})$

detonation which is indicated by a high luminosity
and the occurence of the retonation wave. The ratio
of the initiation distance over the initiation time,
the Oppenheim velocity, divided by the velocity of
sound of the undisturbed gas was plotted against the
fuel gas concentration (Fig. 14). The extrapolation
of these curves to $M_{Opp} = \dfrac{V_{Opp}}{a_o} \longrightarrow 0$ gives the
limits of detonability, measured in long tubes of
the same diameter with the experimental designs described
above. This approach is a very simple one to determine
the limits of detonability. It is at least a .ery good
method to estimate the limits of detonability, which
can otherwise only be measured with great experimental
expenditure.

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13. ABSTRACT

The influence of initial temperature on the limits of detonability of gaseous detonations is investigated for mixtures of CH_4-O_2, H_2-O_2, and H_2-air at one atmosphere initial pressure in long tubes of 4,6,10,16, and 26mm, inner diameter, respectively, by use of a rotating drum camera. Two experimental designs are described to measure these limits at low initial temperatures. The regime of fuel gas concentrations for stable detonations becomes somewhat narrower at lower initial temperatures. The influence of the initial temperature on the critical tube diameter is estimated. Furthermore the applicability of the Chapman-Jouguet theory is discussed in relation to shock wave configurations, i.e. the Mach triple configuration. The essential arguments for the existence of the limits of detonability are discussed. Finally a new method to estimate the limits of detonability with a very simple experimental arrangement is described. By this method experiments concerning the stability of a detonation and its initiation process are linked together.

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14. KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Initial temperature						
Limits of detonability						
Chapman-Jouguet theory						
Shock wave configurations						
Critical tube diameter						
Onset of a detonation						
Tube length						